

Fermi Surface as the Driving Mechanism for Helical Antiferromagnetic Ordering in Gd-Y Alloys

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The first direct experimental evidence for the Fermi surface (FS) driving the helical antiferromagnetic ordering in a gadolinium-yttrium alloy is reported. The presence of a FS sheet capable of nesting is revealed, and the nesting vector associated with the sheet is found to be in excellent agreement with the periodicity of the helical ordering.

A considerable body of theoretical and indirect experimental evidence indicates that the geometry of the Fermi surface (FS) drives a variety of ordering phenomena ; these include exotic magnetic ordering in the rare earths and their alloys [1], compositional ordering concentration density waves in binary alloys [2], and magneto-oscillatory coupling in magnetic multilayers separated by non-magnetic spacer layers [3]. Current theoretical understanding suggests that the ordering is governed by the “nesting” of specific sheets of FS in the disordered state. Nesting describes the coincidence of two approximately parallel FS sheets when translated by some distance in \mathbf{k} -space (i.e. by the “nesting vector”, \mathbf{Q}). In the presence of nesting, the disordered phase becomes unstable to an ordering modulation whose period is inversely proportional to the relevant nesting vector. However, in many cases the relevant features of the FS have never been directly observed.

Despite intense current interest in the concept of FS nesting as the driving force for modulating magnetic structures, there is a distinct dearth of direct experimental information about FS topologies in the relevant materials. This scarcity has mainly been due to the lack of a suitable technique. With recent developments in positron annihilation Fermiology (see e.g. [4]), such studies are now possible. In this Letter, we provide the first direct evidence that a Gd-Y alloy which orders antiferromagnetically does indeed contain a FS sheet that has nesting properties. We compare the nesting vector directly calipered from the FS topology with those inferred via neutron diffraction on specimens from the same batch. We also provide preliminary discussion of our results in terms of the temperature dependence of the nesting vector in the helical phase and effects of the transitions on

this nested FS.

A classic example of such a FS-driven ordering is the helical antiferromagnetic ordering in many of the heavier rare earths (e.g. Tb, Dy, Ho, Er). Here, the magnetic moments align in the basal planes with a rotation of the moment vectors in successive planes with a periodicity that is predicted to arise from the FS topology. The ordering has its origin in the coupling of the localised $4f$ moments via the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange interaction involving the conduction electrons [1]. The connection between the conduction electrons’ ability to establish magnetic order and the FS of the disordered paramagnetic state is most easily understood in terms of the wave-vector (\mathbf{q}) dependent susceptibility,

$$\chi(\mathbf{q}) \propto \sum_{\mathbf{k}, j, j'} \frac{|M(\mathbf{k}, \mathbf{k} + \mathbf{q})|^2 f_{kj}(1 - f_{k+\mathbf{q}+\mathbf{G}j'})}{\epsilon_{j'}(\mathbf{k} + \mathbf{q} + \mathbf{G}) - \epsilon_j(\mathbf{k})}. \quad (1)$$

Here M are the matrix elements involving the conduction and f electron wave functions, f_{kj} are the Fermi-Dirac distribution functions for reduced wave-vector \mathbf{k} and band j , $\epsilon_j(\mathbf{k})$ are the single particle energies and \mathbf{G} is a reciprocal lattice vector. Subject to other constraints, the maximum in $\chi(\mathbf{q})$ determines the most stable magnetic structure. If the maximum in $\chi(\mathbf{q})$ occurs at $\mathbf{q} = 0$, the material orders ferromagnetically. If the maximum occurs at a non-zero $\mathbf{q} = \mathbf{Q}$, then a more complex arrangement of the magnetic moments, such as helical antiferromagnetic order, takes place. The latter may happen if there are large parallel sections of FS to guarantee a sufficient number of terms in the sum with vanishingly small denominators at the nesting vector \mathbf{Q} . A so-called “webbing” feature [5–7] in the FS of most of the rare earths provides the required parallel surfaces for nesting which drives the magnetic ordering.

Helical antiferromagnetic ordering is also observed in Gd-Y alloys in a certain composition and temperature range. The Gd FS does not contain the webbing feature [8] and Gd orders ferromagnetically below 293K. The transition metal Y, on the other hand, possesses a strong webbing sheet [5–7] but owing to the lack of magnetic moments is a paramagnet at all temperatures. Addition of small amounts (of the order of 0.5 at.%) of Tb [10,11]

or Er [12] leads to the appearance of long range magnetic structures with \mathbf{Q} vectors close to the nesting vector in the webbing feature in Y. The helical phase observed in a range of Gd-rich Gd-Y alloys is assumed to arise from a combination of magnetic moments contributed by Gd and the Y-induced nesting character of the FS. Gd-Y provides an ideal alloy system to study the generic magnetic behaviour in the rare earths because of the availability of good quality single crystal samples of sufficient size. Further, the concentration versus temperature magnetic phase diagram in these alloys is well established and is rich in interesting features, many of which are common to other rare earths [13,14]. In the concentration range of 30–40 at.% Y, the alloy shows a helical phase where the helix periodicity, often quoted as the interlayer turn angle of the basal plane moment vector, shows a reversible decrease with temperature. This may imply a temperature-dependent nesting vector (i.e. T -dependent changes in the webbing FS sheet). In the concentration-temperature regime which supports the antiferromagnetism, the application of a modest magnetic field (a few times 10^{-2} tesla) along the c -axis leads to a ferromagnetic alignment of the moments along this axis. The specimen reverts to its antiferromagnetic state once the magnetic field is switched off. In addition to the suggestion that the Fermi surface of the magnetically disordered paramagnetic phase drives the helical ordering, there is the issue of the effect of helical ordering on the electron energy bands and therefore the impact on the FS itself.

The measurements of the FS topology were conducted via the so-called 2-Dimensional Angular Correlation of electron-positron Annihilation Radiation (2D-ACAR). A 2D-ACAR measurement yields a 2D projection (integration over one dimension) of the underlying two-photon momentum density, $\rho(\mathbf{p})$. Within the independent particle model,

$$\begin{aligned}\rho(\mathbf{p}) &= \sum_{\mathbf{k},j} \left| \int d\mathbf{r} \psi_{\mathbf{k},j}(\mathbf{r}) \psi_+(\mathbf{r}) \exp(-i\mathbf{p}\cdot\mathbf{r}) \right|^2 \\ &= \sum_{j,\mathbf{k},\mathbf{G}} n^j(\mathbf{k}) |C_{\mathbf{G},j}(\mathbf{k})|^2 \delta(\mathbf{p} - \mathbf{k} - \mathbf{G}),\end{aligned}\quad (2)$$

where $\psi_{\mathbf{k},j}(\mathbf{r})$ and $\psi_+(\mathbf{r})$ are the electron and positron wave functions, respectively, and $n^j(\mathbf{k})$ is the electron occupation density in \mathbf{k} -space in the j^{th} band. The $C_{\mathbf{G},j}(\mathbf{k})$ are the Fourier coefficients of the electron-positron wave function product and the delta function expresses the conservation of crystal momentum. $\rho(\mathbf{p})$ contains information about the occupied electron states and their momentum $\mathbf{p} = \hbar(\mathbf{k} + \mathbf{G})$ and the FS is reflected in the discontinuity in this occupancy at the Fermi momentum $\mathbf{p}_F = \hbar(\mathbf{k}_F + \mathbf{G})$. As already pointed out, the 2D-ACAR spectra represent projections of $\rho(\mathbf{p})$, and the full 3D density can be reconstructed using tomographic techniques from a small number of projections with integrations along different crystallographic directions [15–17].

Finally, if the effects of the positron wave function (Eq. 2) are small such that the $C_{\mathbf{G},j}(\mathbf{k})$ are almost independent of \mathbf{k} , the full 3D \mathbf{k} -space occupation density can be obtained by folding back the 3D $\rho(\mathbf{p})$ into the first Brillouin zone (BZ) according to the so-called LCW prescription [18]. By these means one is able to directly ‘image’ the FS in its full 3D form.

The sample under investigation was a $\text{Gd}_{0.62}\text{Y}_{0.38}$ single crystal which undergoes a helical antiferromagnetic transition below $\sim 200\text{K}$ where the nature of the helical phase and its periodicity has been extensively studied by one of us [14]. As part of a comprehensive programme, we have also studied the FS topology of the two pure elements Gd [8,9] and Y [7,9]. In each case, the 3D $\rho(\mathbf{p})$ and subsequently the 3D \mathbf{k} -space occupancy were reconstructed from five projections measured at 7.5 degree intervals encompassing the 30 degrees between the directions Γ -M and Γ -K (in the irreducible wedge of the hexagonal BZ). Following the usual processing of the measured 2D-ACAR spectra [4], they were deconvoluted using a ‘Maximum Entropy’-based (MaxEnt) procedure [19] to suppress the unwanted smearing due to experimental resolution. The 3D $\rho(\mathbf{p})$ was reconstructed from the measured projections (both raw and deconvoluted, henceforth referred to as ‘raw’ and ‘MaxEnt’) using Cormack’s method [15–17] before finally being folded back into the first BZ. The reconstruction method exploits the crystallographic symmetry which allows the reconstruction from only a few projections and has been rigorously tested [17,9] to show that no artefacts are introduced into the data. A more detailed description of the procedures used here can be found in [17] and further references therein. Using a threshold criterion to differentiate between the empty and occupied states [20,7] it is possible to image the FS alone.

In Figure 1, we show FS images of: (a) the calculated FS for Y [5], where the webbing feature and the associated nesting vector, \mathbf{Q}_0 , is marked by the double arrow; (b) the measured Fermi surface of Y [7] clearly showing the webbing feature; (c) the measured FS of Gd [8,9] with the distinct lack of the webbing feature as expected from its ferromagnetic ordering (d) the current measurement of the alloy $\text{Gd}_{0.62}\text{Y}_{0.38}$ (in the disordered paramagnetic phase) where the nesting feature is clearly visible. The presented experimental FS images are extracted from MaxEnt deconvoluted data. It is noteworthy that in the paramagnetic phase of the Gd-rich alloy, the webbing feature is remarkably similar to that observed in pure Y. The fact that the FS of pure Gd does not show the webbing feature while that of the alloy has a strong nesting character clearly indicates that its helical ordering is driven by the nesting of this sheet.

We now investigate the webbing in greater detail. Figure 2 shows the section through the webbing in the alloy sample (Fig. 1d) in the H-L-M-K plane (on the face of the BZ in Fig. 1a). Here, lows (holes) are shown as black and

highs (electrons) as white and the webbing is represented by the central region of holes. If the raw and Maxent reconstructions are normalised to contain the same number of electrons within the BZ and the raw reconstruction is subtracted from the Maxent reconstruction, the distribution shown in Figure 3 results. This procedure amounts to an edge-enhancement that highlights the Fermi edges [19,9]. It enhances the discontinuity at the FS because it is in the vicinity of these discontinuities that the resolution function has its greatest smearing effect. Thus, the difference spectrum amplifies the signature of the Fermi edge.

Dugdale *et al.* [19] proposed that the locus of points where such a difference spectrum passes through zero defines the Fermi surface, providing an accurate method for calipering it. The zero crossing contour of Figure 3 is shown in Figure 4 which again clearly shows the yttrium-like webbing feature and its nesting properties. We estimate the width of the webbing parallel to the c -axis as $0.53 \pm 0.02 \times (\frac{\pi}{c})$, a value remarkably close to the nesting feature in pure Y of $0.55 \pm 0.02 \times (\frac{\pi}{c})$ [7]. This would give rise to a period of helicity which corresponds to an inter-plane turn angle of 47.7 ± 1.6 degrees between the orientations of the magnetic moments in successive basal planes. As mentioned earlier, Bates *et al.* [13] measured the turn angle in the helical phase in a sample derived from the same ingot via neutron diffraction by inspecting the distance between the magnetic satellite Bragg peaks on either side of the nuclear peak along the c -axis. These results showed that the measured turn angles were strongly T -dependent, increasing linearly with temperature. The turn angles cannot be measured in the paramagnetic phase through neutron diffraction owing to the absence of the magnetic ordering and therefore the lack of the satellite peaks. However, a linear interpolation of the T -dependence of the turn angle returns a value of 48 degrees at 295K (the temperature of our measurement) which is in excellent agreement with the value obtained in our experiment.

Finally, it is worth mentioning our recent preliminary results from the same alloy sample measured at $T = 140\text{K}$, well within the thermodynamic helical phase. In our experimental set-up, we use a magnetic field of $\sim 0.8\text{T}$ to focus the positrons on to the specimen. The field is applied parallel to the c -direction of the sample. It was not possible to carry out the measurements without the magnetic field owing to the small dimensions of the sample, as a substantial fraction of the defocused positrons annihilated in the sample holder and associated goniometer. As noted above, the applied field would force the magnetic moments to align along the easy axis giving rise to a c -axis ferromagnetic state. Our preliminary analysis of the FS of the alloy sample in this ferromagnetic state reconstructed from 3 projections again shows (figure not shown owing to lack of space) distinct indica-

tion of the webbing feature being present.

If the periodic antiferromagnetic ordering introduced superzone boundaries in the lattice, these may have distorted the FS feature in the webbing sheet. However, in our low temperature data, such degeneracy would have been lifted by the forced ferromagnetic ordering. If this is the case and the relevant energy bands are not significantly affected, then one would expect the webbing feature in the FS to be retained. Although theoretical calculations are necessary to confirm this, it is a reasonable assumption.

In conclusion, we have shown for the first time the existence of a nesting FS in a Gd-Y alloy ; this confirms theoretical predictions that the FS topology in the paramagnetic state is responsible for the antiferromagnetic ordering of the alloy.

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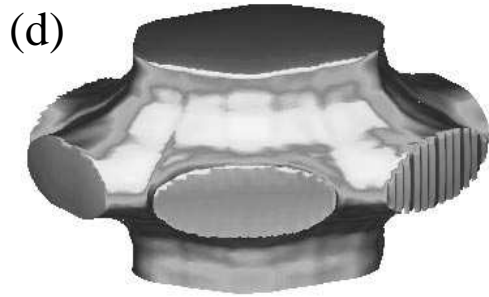
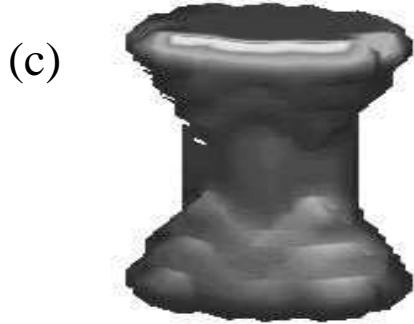
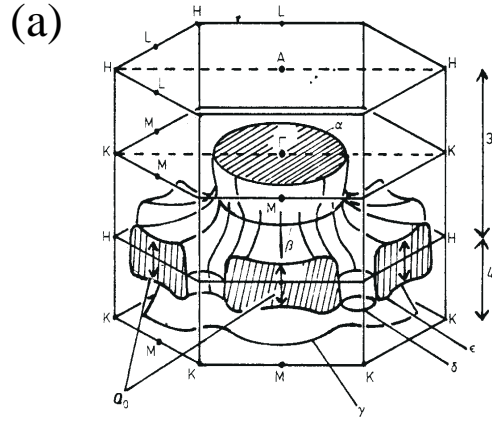


FIG. 1. (a) Calculated FS of Y [5]. (b) Measured FS of Y ; note the presence of the ‘webbing’ feature. (c) Measured FS of Gd ; note that there is no webbing. (d) Measured FS of $Gd_{0.62}Y_{0.38}$, showing a webbing feature.

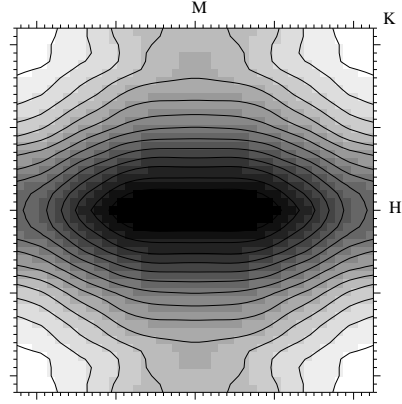


FIG. 2. Electron density in H-L-M-K plane. Black signifies holes, and white represents electrons.

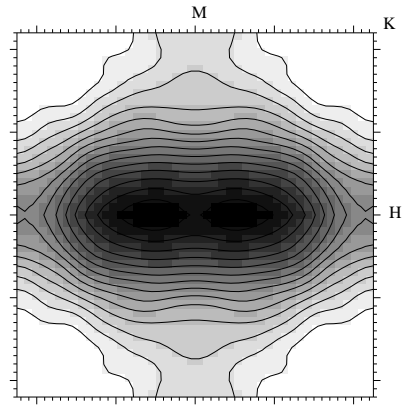


FIG. 3. Difference between MaxEnt [19] and raw electron densities in H-L-M-K plane.

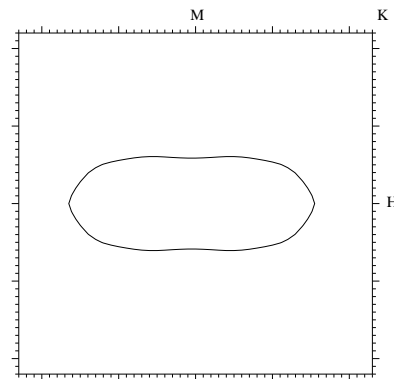


FIG. 4. The ‘zero contour’ of Fig.3. Dugdale *et al.* showed that this indicates the Fermi surface.